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Resonant Plasmonic Enhancement of InGaN/GaN LED using Periodically Structured Ag Nanodisks

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Abstract: Ag nanodisks are fabricated on GaN-based LED to enhance emission efficiency. Nanosphere lithography is used to obtain a periodic nano-structure, and a photoluminescence enhancement of 2.7 is reported with Ag nanodisk diameter of 330 nm.

OCIS codes: (230.3670) Light-emitting diodes; (240.6680) Surface plasmons

1. Introduction

GaN-based light-emitting diodes (LEDs) have shown a promising role in the search for energy efficient visible light sources. By incorporating indium (In), the bandgap of InGaN can be tuned from near UV to over the entire visible spectrum. LEDs based on III-arsenide and phosphide material systems already constitute efficient light sources from the red to yellow spectral region, thus III-nitrides could play a crucial role for LEDs from blue to green. The internal quantum efficiency (IQE) of blue InGaN/GaN LED grown by metalorganic chemical vapor deposition (MOCVD) on sapphire substrate, easily reaches above 80 %. However, in moving towards longer wavelengths by increasing the In composition, the IQE suffers a significant decrease, resulting in a poor performance of green LEDs [1].

One approach to improve the IQE of LEDs, which has been heavily researched over the last couple of years, involves the fabrication of Ag nanoparticles (NPs) in the near field of the active light emitting region, where the coupling between excitons and localized surface plasmon (LSP) modes is expected to provide a faster radiative decay channel [2]. The simplest fabrication approach is self-assembled Ag NPs by thermal annealing [3]. However, because of the difficulty in controlling the particle sizes, this method allows limited control over the LSP resonance.

Here we propose a simple fabrication method of Ag nanodisks (NDs) with a periodic pattern based on nanosphere lithography (NSL), with which both the particle size and the periodicity can be controlled. While the resonance position can be tuned with the size, the periodicity allows control over the resonance width [4].

2. Fabrication Process

All the samples were prepared on a multi-quantum well (MQW) InGaN/GaN green LED grown by MOCVD on a *c*-plane sapphire substrate. In order to have QW-LSP coupling the thickness of the Mg-doped p-GaN layer was 27 nm. The process flow of fabricating a periodic structure of Ag NDs using NSL is illustrated in Fig. 1. First two thin layers of Si₃N₄ and SiO₂, for pattern reversal, were deposited by plasma enhanced chemical vapor deposition (PECVD), where SiO₂ provides an adhesion layer for the dielectric nanospheres and Si₃N₄ is a passivation layer for GaN surface before SiO₂ deposition.

Polystyrene nanospheres with a diameter of 595 ± 5 nm were deposited on the surface via the Langmuir-Blodgett technique to form a colloidal structure of NPs, Fig. 1(a). The success of this step was very dependent on temperature, humidity, applied pressure, and the aqueous solution concentration of the nanospheres.

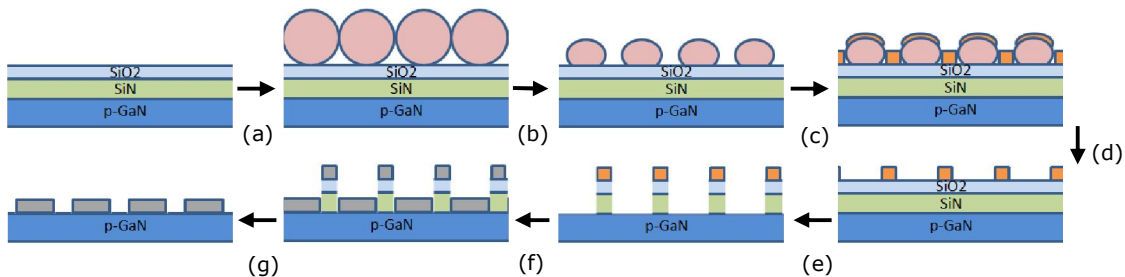


Figure 1: Process flow.

The diameter of these spheres determines the periodicity (pitch) of Ag NDs. However, to control the size, an O_2 reactive ion-etching (RIE) process was employed to reduce the size of the polystyrene nanospheres, Fig. 1(b). The corresponding scanning electron microscope (SEM) image after O_2 RIE is shown in Fig. 2(a).

As an etching-mask, a 30 nm layer of Ni was deposited by e-beam evaporation, Fig. 1(c), whereafter the nanospheres were removed by sonication in acetone, Fig. 1(d). The pattern formed by the nanospheres was transferred to the Si_3N_4/SiO_2 layer by an RIE process, Fig. 1(e), and the Ni layer was then removed by an aqua regia solution. A 30 nm layer of Ag was deposited, Fig. 1(f), and a lift-off process was done through wet etching the Si_3N_4/SiO_2 with BHF, leaving behind a periodic pattern of Ag corresponding to that of the etched nanosphere pattern, Fig. 1(g). The Ag NPs, however, had the shape of a disk with a diameter equal to that of the etched nanospheres, as shown in Fig. 2(b), and a thickness of 30 nm.

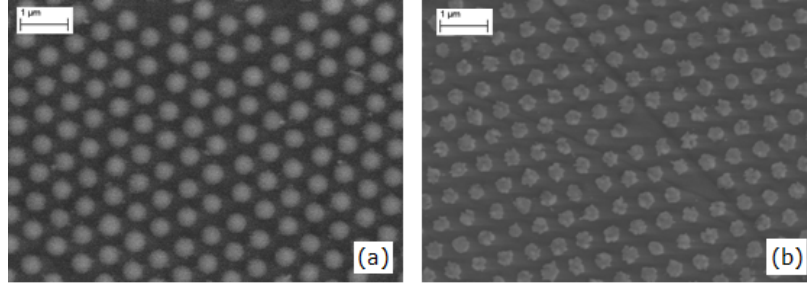


Figure 2: SEM image of (a) reduced-size colloidal nanospheres, and (b) the resulting Ag ND formation.

3. Optical Characterizations

Four samples were prepared with different Ag ND average diameters, corresponding to 380, 330, 240 and 130 nm. The photoluminescence (PL) spectra were measured using a backside excitation and detection setup. The excitation laser had a wavelength of 375 nm and an excitation power of 1 mW. PL spectra of the 380 and 330 nm samples are shown in Fig. 3(a). The difference in the as-grown PL intensity and peak wavelength is due to variations of In composition across the wafer. Therefore, reference is always taken along contours of equal wavelength. The peak around 425 nm is from the InGaN/GaN super-lattice (SL) structure of the LED which is grown before the MQWs. It is noticed that the intensity at 425 nm is almost unaffected by the Ag NDs for both samples, which is due to large spacing between p-GaN surface and the SL structure, ~ 100 nm, leading to an inefficient coupling from excitons to LSP mode at the blue wavelength. This would indicate that the enhanced intensity at the green wavelength is due to MQW-LSP coupling which provides a faster radiative decay channel for the excitons and increases the IQE. Although the as-grown PL spectra for 380 nm and 330 nm samples are different, they are same for the 330, 240 and 130 nm samples as shown in the inset of Fig. 3(a).

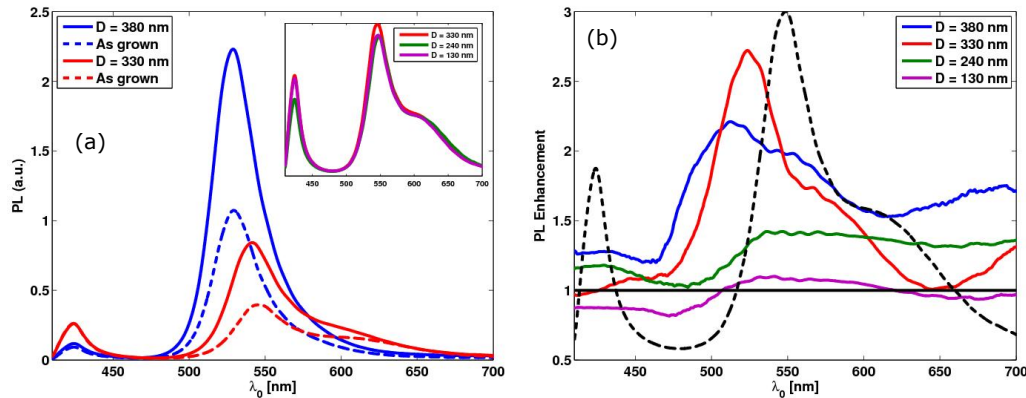


Figure 3: (a) The PL spectra of two samples, comparing areas with (solid) and without (dashed) Ag nanodisks. The inset shows the remaining as-grown PL spectra. (b) PL enhancement factors for samples with different Ag disk diameters. The dashed line is the as-grown emission spectrum of the 330, 240 and 130 nm samples.

The PL enhancement factor is shown in Fig. 3(b). The sample with Ag NDs of 330 nm diameter has the highest enhancement factor of about 2.7 at a wavelength of 523 nm, while the as-grown emission peak wavelength is around 545 nm. Considering the 380 nm sample, the peak enhancement factor is about 2.2 around 513 nm. The 240 nm

sample appears to have a very broad enhancement despite being below a factor of 1.5, while at the same time the sample with the smallest diameter has the lowest enhancement factor.

However, when considering the LSP resonance through a reflectance measurement, as shown in Fig. 4(a), the 130 nm sample demonstrates the most pronounced resonance, although very weak as seen by the absorption spectra in Fig. 4(b). Absorption is obtained by $A = 1 - T - R$, where T and R are the transmittance and reflectance, respectively. From the absorption spectrum of the 240 nm sample, it would appear that an LSP resonance could be located in the long wavelength region. Thus we might understand how the enhancement factor can have such a flat shape as in Fig. 3(b), if we assume that the PL enhancement follows the LSP resonance.

Except for the 380 nm sample, the enhancement factors follow the trend of the absorption strengths. For instance, while the 130 nm sample has the smallest absorption it also has the smallest enhancement. The decrease in absorption is well correlated with the decrease in metal surface coverage as the diameters are decreased while the lattice periodicity remains constant. The reflectance spectra suggest that the increasing PL enhancement with metal surface coverage is not due to reflection. Measurement of the transmittance spectra includes both the specular and diffusive transmission using an integrating sphere. Therefore, the significant increase in the quantity $1 - T - R$ is due to absorption as opposed to scattering, since the latter is already included in the quantity, T .

The observations show that decreasing the particle size while keeping the pitch fixed, results in a reduced field-enhancement effect. This suggests that the dipole-dipole interaction is weakened. Similar effect has been observed for disordered nanoparticles, where the interaction is significantly reduced for pitch-to-size ratios larger than 2 [5]. Thus to keep the advantage of a periodic structure the Ag NDs must keep their close proximity, and the pitch should be reduced together with the particle size, keeping in mind that an optimum pitch exists for a given particle size [4].

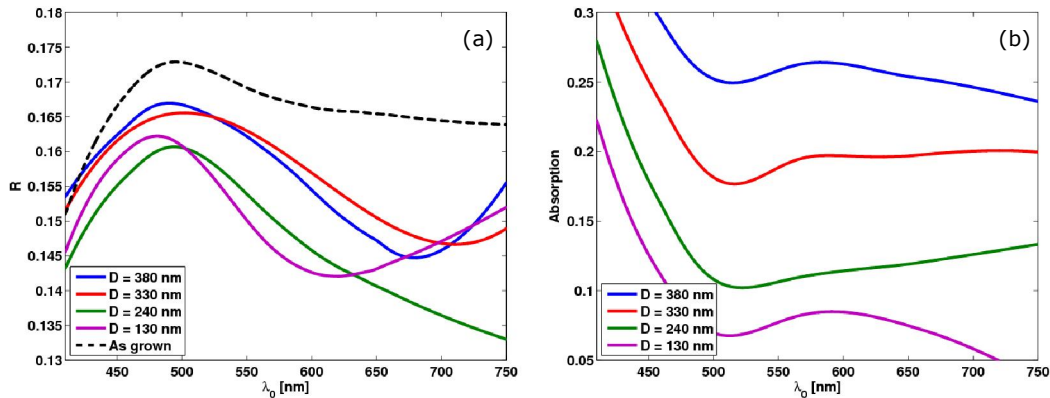


Figure 4: (a) Reflectance and (b) absorption spectra of the various diameters of Ag NDs.

4. Conclusion

A periodic pattern of disk-shaped Ag NPs were fabricated using NSL together with the Langmuir-Blodgett deposition technique of dielectric nanospheres serving as the lithography pattern template. The pitch and diameter of the final Ag NDs could be controlled by the initial and post-etch diameter of these nanospheres. PL characterizations revealed an enhancement by a factor of 2.7 for the Ag ND diameter of 330 nm. Decreasing the Ag ND size below 240 nm, while keeping the center-to-center distance at 600 nm, has shown to be ineffective for PL enhancement. To obtain a smaller pitch, smaller dielectric nanospheres need to be used as templates during the NSL process.

5. References

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